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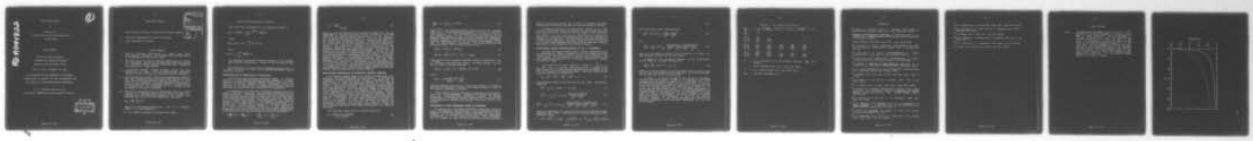

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SOLID STATE OPTICS

by

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Final Report

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SOLID STATE OPTICS

This contract concerned itself with two major topics:

1 Nonlinear Interaction of Light with Matter

2 Laser Operation and Design

BRIEF SUMMARY

1.1 Work on nonlinear interaction of light with antiferromagnetic materials, and thermoelastic materials ~~by H.~~ Ho is described in more detail below.

1.2 The Dirac Theory of Poisson brackets usually used to handle the constraint problem in electrodynamics has been applied by C. Valenti to handle magnetic materials whose magnetization vector has a fixed length.

1.3 A microscopic approach toward nonlinear optics has been started, by C. Huang. His work reported here, provides a localized orbital estimate for nonlinear optical constants.

2.1 The paraxial wave optics[1] method with intensity and slope as the key variables avoids the rapid phase fluctuations that occur when field amplitudes are used as variables. A program to use equations for these variables to describe an unstable resonator with rectangular mirrors is being written in collabor tion with Professor W. H. Louisell of the University of Southern California.

2.2 The spatial distribution of the temperature rise in a solid induced by a Gaussian laser beam of width w and power P is expressed as a one dimensional integral. The maximum temperature rise is given by the closed form expression

$$T_{max} = \frac{P}{2Kw} \frac{1}{\sqrt{\pi}} N(W)$$

where K is the thermal conductivity, N(W) is a reduction factor (< 1) that depends on

$W = \alpha w$ = (power attenuation constant)(beam width)

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= (beam width)/(attenuation length).

The closed form expression for the reduction factor is

$$N(W) = WD(\tfrac{1}{2}W) - \frac{W}{2\sqrt{\pi}}\, e^{-\frac{1}{4}W^2}\, Ei(\tfrac{1}{4}W^2)$$

where

$$D(x) \equiv \exp(-x^2) \int_0^{\infty} \exp t^2\, dt$$

and

$$Ei(x) = P \int_{-\infty}^{\infty} \frac{\exp(t)}{t}\, dt$$

the tabulated functions[2], Dawson's integral and the exponential integral respectively. A plot of N(W) is given in Fig. 1.

As an example, a 5μ beam with an attenuation length $1/\alpha \simeq 1\mu$ in GaAs has $W = 5$, $N = .830$ and a maximum temperature rise of $(124)(.83) = 103^{\circ}K$.

Electrodynamics in Thermoelastic Dielectrics

An ab initio long wavelength Lagrangian theory of electrodynamics of thermoelastic dielectrics was developed. It's purpose is to extend the Lagrangian method developed by Lax and Nelson[4,5,6] to include thermal effects. In general, this is the treatment of the interactions of electromagnetic fields, acoustic waves and heat conduction with dissipation in dielectrics.

Our approach is to construct a Lagrangian density which consists of the field Lagrangian, the interaction Lagrangian and the matter Lagrangian. The matter Lagrangian is equal to the kinetic energy less the internal energy U which does not arise from classical electromagnetic forces. The internal energy is a function of the finite strain tensor, body components of the excitation fields and the entropy density S. The absolute temperature T is defined by $T \equiv \partial U/\partial S$, and in general is a function of the position vectors. The free energy density F is then defined by $F \equiv U - TS$, so F is a function of the finite strain tensor, body components of the excitation fields and the absolute temperature. By the definition of the free energy, we have the relations:

$$\left(\frac{\partial U}{\partial E_{AB}}\right)_S = \left(\frac{\partial F}{\partial E_{AB}}\right)_T , \quad \left(\frac{\partial U}{\partial y_i^{\mu}}\right)_S = \left(\frac{\partial F}{\partial y_i^{\mu}}\right)_T$$

$$S = -\left(\frac{\partial F}{\partial T}\right)_{E_{AB}, y_i^{\mu}} \tag{1}$$

where E_{AB} are the components of finite strain tensor and y_i^{μ} are components of internal excitation fields. Therefore, the stress tensor and the thermal expansion force acting on the particles can be derived from the free energy F. As far as the irreversible processes are concerned, there is a friction force which can not arise from the potential energy. A dissipation function D can be defined[7,8] by the rate change of the internal entropy production. The dissipation function D is a function of the rate change of deformation, the rate change of body components of the excitation fields and the temperature gradient. All equations of motions for the fields and matter with thermal effects and the constitutive equation of Duhamel-Neumann Law[9] can be derived from the Lagrangian and the dissipation function without a phenomenological formulation. The conservation laws of momentum, angular momentum and energy are automatically satisfied from the field equations and the invariance properties of the free energy F and the dissipation function D. The equation of heat conduction coupled with deformation and excitation fields is also discussed. In further work we will specialize to linear thermolasticity and piezoelectricity and intend to apply these results to ferromagnetics, antiferromagnetics and liquid crystals.

Photon-Magnon Interaction in Anisotropic Magnetic Crystals

The coupling between photons and magnons in anisotropic ferromagnets and antiferromagnets has been studied. By solving the equations of magnetic field $\bar{H}$ from Maxwell's equations and of magnetization $\bar{M}$ under the effects of constant and rf fields, we shall be able to obtain the dispersion relation of the coupled system in anisotropic crystals. When the proper boundary conditions are applied, we can obtain the coefficients of reflectivity and transmissivity. Both coefficients depend on the external static magnetic field $\bar{H}_0$ and the thickness of the film. The possible application of using a thin-film antiferromagnetic crystal as an optical device to obtain pure circularly polarized radiation by transmission has been discussed before by Zuniga and Bose[10,11]. The Green's function method developed by Lax and Nelson[4,12] was applied to solve the magnetic field equation in anisotropic crystals. The results were used to obtain the absorption or scattering power which can be applied to study the spin resonance spectra in anisotropic ferromagnets and antiferromagnets.

Our treatment starts from the coupled equations:

$$\bar{H} = \frac{-\bar{K}(\bar{K} \cdot \bar{M}) + \omega^2/c^2\, \bar{M}}{K^2 - \omega^2/c^2} \tag{2}$$

$$-\frac{d\bar{M}}{dt} = \gamma \bar{M} \times [\bar{H}_0 + \bar{H} + \bar{H}^{eff}] \quad , \tag{3}$$

where $\bar{H}^{eff}$ is the effective crystal field and γ is the gyromagnetic ratio. The abbreviated notations $\bar{k} = -i\bar{\nabla}$ and $\omega = i\partial/\partial t$ are used to get Eq. (2). The magnetization can be solved in terms of the magnetic field, $\bar{M} = \underline{x}_m \cdot \bar{H}$, using Eq. (3), where $\underline{x}_m$ is the magnetic susceptibility tensor which is a function of frequency, constant magnetic field $\bar{H}_0$ and anisotropy field of the crystal. M can be substituted into Eq. (2) to obtain the dispersion relation of photon-magnon interaction. If we are looking for the scattering power, Eq. (2) can be rewritten as

$$\underline{\alpha}(\bar{k},\omega) \cdot \bar{H}(\bar{r},\omega) = \bar{M}^{NL}(\bar{r},\omega) \tag{4}$$

$$\underline{\alpha}(\bar{k},\omega) = (\frac{c}{\omega})^2 [\, k^2 \underline{1} - \bar{k}\,\bar{k}\,] - \underline{K}_m(\omega) \tag{5}$$

where $\underline{K}_m(\omega)$ is the frequency dependent magnetic permeability tensor $\bar{M}^{NL}(\bar{r},\omega)$ is the nonlinear part of the magnetization. Eq. (4) has the Green's function solution[3]

$$\bar{H}(\bar{r},\omega) = \int \underline{G}(\bar{r} - \bar{r}') \cdot \bar{M}^{NL}(\bar{r}')\, d\bar{r}'\, e^{-i\omega t} \quad , \tag{6}$$

where

$$\underline{G}(\bar{R}) = \int \frac{\exp(i\bar{k} \cdot \bar{R})}{\underline{\alpha}(\bar{k},\omega)} \frac{d\bar{k}}{(2\pi)^3} \quad , \tag{7}$$

When the magnetic field in Eq. (7) has been solved, it is easy to calculate the electric field and the scattering power in terms of the magnetic permeability tensors.

These results obtained here will be applied to the ferromagnetic and antiferromagnetic crystals with uniaxial and biaxial structures. In further work, the photons, magnons and photons interaction in anisotropic magnetic crystals will be studied, and the interaction of magnons with electric field will also be considered.

Application of Dirac Constraint Theory to Magnetism

In working out a consistent macroscopic Lagrangian theory of linear and nonlinear electrodynamics for an anisotropic magnetic dielectric possessing acoustic, ionic, electronic and spin excitations, it is necessary to identify the classical coordinates corresponding to the spin degrees of freedom. This identification is needed in order to obtain the Euler-Lagrange equation

relating to the spin motion, and so that a complete canonical stress tensor containing the spin contributions may be derived.

A classical Lagrangian and Hamiltonian theory of a rigid magnetic continuum, easily generalized to the elastic case, has been formulated which indicates that the choice of the magnetization $\bar{m}(\bar{z},t)$ and its time rate of change $\dot{\bar{m}}(z,t)$ as generalized coordinates and velocities corresponding to the spin degrees of freedom, is a proper one. This calculation also arouses interest in that Dirac's generalized dynamics, hitherto used in connection with quantum electrodynamics and quantum field theories, finds an application in the physics of the continuum solid.

Second Order Optical Susceptibilities of III - V Compounds

The long wave length second order optical susceptibilities of tetrahedrally bonded materials can be calculated within the context of the bond orbital model[3], which is an empirical tight binding model. By the symmetry of the zinc blende structure, the only nonvanishing components of the second order optical susceptibilities is $x^{(2)}_{123}$.

The electrons will gain a potential energy $U = \sum_{i=1}^{3} e E_i x_i$ per bond when an external electric field $\bar{e}$ is applied to the solid, where x_i is the coordinate measured from the center of the bond and is along the three cubic edges. The ground state energy E_G of one bond can be expanded as

$$E_G = E_G^{(0)} + E_G^{(1)} + E_G^{(2)} + E_G^{(3)} + \cdots \quad . \tag{8}$$

Each expanding terms can be expressed by the usual perturbation theory as

$$E_G^{(1)} = \sum_i E_i \langle G|X_i|G\rangle = e E_i \bar{X}_i \quad , \tag{9}$$

$$E_G^{(2)} = - \sum_{i,j} e^2 E_i E_j \sum_M{}' \frac{\langle G|X_i|M\rangle \langle M|X_j|G\rangle}{E_M^{(0)} - E_G^{(0)}} \quad , \tag{10}$$

$$E_G^{(3)} = \sum_{ijk} e^3 E_i E_j E_k \sum_{L,M}{}' \frac{\langle G|X_i|L\rangle\langle L|X_j - \bar{X}_j|M\rangle\langle M|X_k|G\rangle}{(E_L^{(0)} - E_G^{(0)})(E_M^{(0)} - E_G^{(0)})} \quad , \tag{11}$$

where the summations of L and M are over all the exciting states. Comparing the above equations with the total macroscopic electrostatic energy:

$$W = NE_G = NE_G^{(0)} - \sum_i P_i E_i - \sum_{i,j} \frac{1}{2} x^{(1)}_{ij} E_i E_j - \frac{1}{3} \sum_{i,j,k} x^{(2)}_{i,j,k} E_i E_j E_k \cdots$$

(12)

one obtains the linear susceptibility,

$$x_{ij}^{(1)} = \frac{2}{3} N|e|^2 \sum_M' \frac{|\langle G|\xi|M\rangle|^2}{E_M^{(0)} - E_G^{(0)}} , \qquad (13)$$

and

$$x_{ijk}^{(2)} = \frac{1}{\sqrt{3}} N|e|^3 \sum_{L,M}' \frac{\langle G|X_i|L\rangle\langle L|X_j - \bar{X}_j|M\rangle\langle M|X_k|G\rangle}{(E_L^{(0)} - E_G^{(0)})(E_M^{(0)} - E_G^{(0)})} , \qquad (14)$$

where N is the number of bonds in unit volume and ξ is along the body diagonal direction of the unit cube.

In terms of the parameters defined in the bond-orbital model[3], $x_{123}^{(2)}$ can be simply expressed as

$$x_{123}^{(2)} = \frac{\sqrt{3}}{2} |e| x^{(1)} d \gamma \alpha_p / E_2 , \qquad (15)$$

where E_2 is the principal optical absorption peak, d is the bond length, α_p is the polarity of the bond and γ is a fitting parameter for each row of the Periodic table.

In table I, the values of $x_{123}^{(2)}$ for all the III - V compounds evaluated here are listed in column I. For comparison the values obtained from the dielectric model of J. C. Phillips and J. A. Van Vechten are listed in column II, the values of D. A. Kleinman using the same model are listed in column III. In column IV and V are the values obtained by the bond charge model of B. F. Levine and the experimental results. Due to the uncertainties in the measurement of the absolute values of $x_{123}^{(2)}$ experimentally the relative values or the trends may be more important in these kinds of simple calculations. The correct trends are obtained here comparing with the experiment as well as the previous authors. However, in the method described here, there are no additional parameters other than those determined in the bond orbital model.

Table I. (In units of 10^{-9} e.s.u)

	I	II (PV)	III (K)	IV (L)	V (Exp)
BN	38	10			
BP	0				
BAs	0				
Al P	363				
Al As	428	1420			
Al Sb	833	1110			
Ga P	524	850	750	260	198
Ga As	716	1220	1080	460	430
Ga Sb	1160	2350	1930	1040	1300
In P	768	1060			
In As	1080	1570	1710	820	860
In Sb	1800	2820	2860	1780	1740

PV : J. C. Phillips & J. A. Van Vechten, Phys Rev. 183, 709 (1969).

K : D. A. Kleinman, Phys. Rev. B 2, 3139 (1970).

L : B. F. Levine, Phys. Rev. B 7, 2600 (1973).

Exp : See also references In L.

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FIGURE CAPTION

Fig. 1 The normalized temperature rise $N(0,0,W)$ at the surface ($z = 0$) and beam center ($r = 0$) for a laser beam of shape $\exp(-r^2/w^2)$ which attenuates as $\exp(-\alpha z)$ plotted versus $W = \alpha w$. The normalization is such that $N(\infty) = 1$, that is, $N(W)$ is the ratio in temperature rise when the heat is deposited in the finite attenuation length $1/\alpha$ to the rise that would be produced if the same heat were absorbed in an infinitesimal layer at the surface. The upper (solid) curve corresponds to the horizontal scale shown. The lower (dashed) curve is the upper curve replotted with the horizontal scale expanded by a factor of 5 to facilitate reading points in the small W region.

Fig. 1